

Computer models reveal the significant effects of aerosols resulting from human activity.

NE of the most controversial scientific issues is determining the causes of the gradual warming of Earth's atmosphere over the past century, especially the last 50 years. Lawrence Livermore scientists have been among the leaders in modeling global climate change to better understand the nature of the warming, to predict the probable climate in the coming decades, and to determine the role of anthropogenic (human) activity in climate change.

Until recently, the most important factor in global climate change appeared to be the steady accumulation of greenhouse gases, mainly produced by the burning of fossil fuels in cars, factories, and power plants. These greenhouse gases, such as carbon dioxide and methane, are known to trap sunlight and thereby warm the atmosphere.

Close observations of global temperature records over the past 50 years have shown less global warming than predicted by computer models that include only accumulations of greenhouse gases. The explanation for this apparent discrepancy is that increasing concentrations of anthropogenic aerosols in the atmosphere may be cooling the planet and so partially counteracting the effects from the greenhouse gases.

In the past 10 to 15 years, scientists have also begun to consider how aerosols, microscopic particles directly suspended in the atmosphere or trapped in clouds, may be changing the planet's climate. Beginning in the early 1990s, calculations showed that aerosols composed of sulfates (a form of sulfuric acid and a main component of air pollution) could be cooling the atmosphere by backscattering incoming solar radiation. The process works in much the same manner as volcanic eruptions, which spew many tons of sulfates into the higher atmosphere that eventually result in the cooling of Earth's climate. (See the box on pp. 10–11.)

Spotlight on Aerosols

In the past few years, intriguing data from ground stations and satellites, together with insight gained from computer models, have made aerosols a major focus of atmospheric research. "Ten years ago, the focus was on greenhouse gases. Now aerosols are getting the attention," says Livermore atmospheric scientist Catherine Chuang.

Chuang notes, however, that large variations in aerosol concentrations have made it difficult to confidently assess the magnitude of their effects on climate. Aerosol chemistry and physics, especially in clouds, are complex and not completely understood. Particles typically remain aloft in the troposphere (lower atmosphere) for a week or less, in contrast to greenhouse gases, which can persist for about a century.

Because they are short-lived, aerosols do not mix homogeneously around the planet's atmosphere, and so concentrations differ greatly from one region to the next. What's more, aerosols come in a wide range of particle sizes, with particles smaller than a micrometer exerting comparatively greater climatic effects. As a result, says Chuang, one of the greatest uncertainties in climatic change is the role played by anthropogenic aerosols. To reduce these uncertainties, scientists are turning to sophisticated computer simulations in an attempt to gain insight into aerosols' climatic effects.

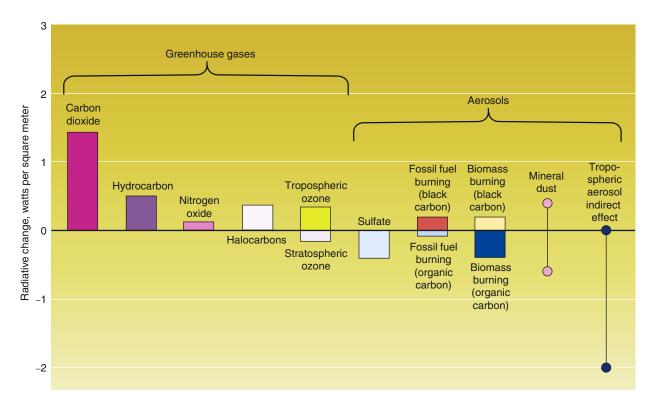
During the past few years, Chuang and colleagues including Joyce Penner (now at the University of Michigan), Keith Grant, Jane Dignon, Peter Connell, Daniel Bergman, and Douglas Rotman have used Livermore's TeraCluster2000 multiparallel supercomputer and the resources of the National Energy Research Scientific Computing Center at Lawrence Berkeley National Laboratory to model how anthropogenic aerosols affect global and regional

climate. The researchers' simulations show in unprecedented detail how aerosols are partially offsetting the effect of global warming and changing the properties of clouds. In some industrial regions, the generation of aerosols from fossil fuel combustion and biomass (forest and grassland) burning may be as important to climate change as greenhouse gases. Also, climate changes caused by aerosols vary significantly by season and by region.

The research team belongs to the Atmospheric Chemistry and Aerosols Group, part of the Atmospheric Science Division of Livermore's Energy and Environment Directorate. The team's advanced simulations, whose findings have been corroborated by field measurements at different geographical locations, build on Livermore's expertise in aerosols, climate, chemistry, and

supercomputer simulations. The research has received funding from the Department of Energy, National Oceanic and Atmospheric Administration, National Aeronautics and Space Administration, and Laboratory Directed Research and Development. The work also contributes to fulfilling the goals of the federal government's National Aerosol Climate Interactions Program, an interagency effort created last year.

Chuang explains that aerosol concentrations from natural sources, such as volcanoes, sea spray, and desert dust storms, are believed to have remained generally steady over the past century. However, like greenhouse gases, anthropogenic aerosols have increased markedly since 1950. Based on satellite data, models, and information on urban and agricultural activities, scientists



Global climate change by greenhouse gases and aerosols since 1750. Factors above zero have a warming effect; those below zero have a cooling effect. A vertical line between two data points indicates scientific uncertainty regarding the estimated contribution of a particular factor.

believe anthropogenic aerosols currently contribute about half of the total submicrometer-size aerosols in the atmosphere. Most of the anthropogenic aerosols are sulfates and carbonaceous compounds produced by the burning of fossil fuels and biomass.

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Solar Reflection Means Cooling

When directly suspended in the atmosphere, most aerosol particles exert a direct cooling effect on the global climate by scattering sunlight back into space. Aerosols also exert a significant indirect effect by serving as cloud condensation nuclei (CCN) for raindrops to form. Increases in CCN result in clouds with more but smaller droplets, thereby increasing the cloud's reflectivity of solar radiation, or albedo. Clouds with

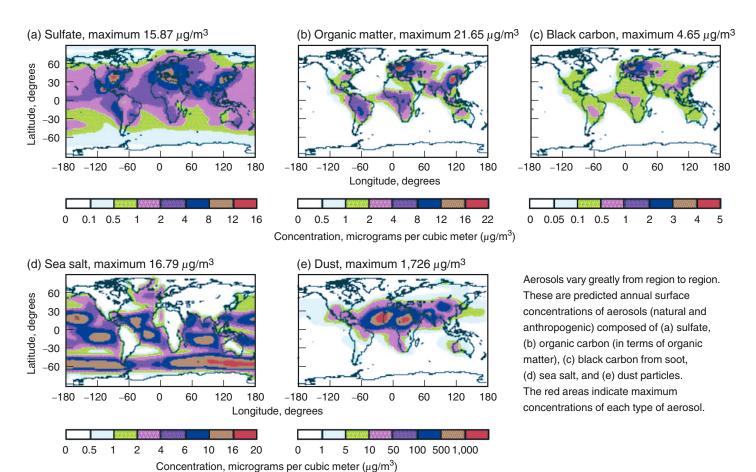
numerous small droplets tend to last longer and so prolong the cooling effect.

Complicating matters is the recently discovered influence of black carbon aerosols, such as soot (incompletely burned carbon), that absorb heat instead of reflecting it back into space. Black carbon aerosols are particularly prevalent over parts of Europe, eastern China, and India, where much coal is burned.

Beginning in the early 1990s, Chuang focused first on modeling the direct effects of anthropogenic sulfate aerosols because they were thought to be the most important compound involved in pollution over China, Europe, and the eastern coast of the United States. She then added the contribution from carbonaceous compounds because of their sizable emission from many industrialized regions of the Northern Hemisphere and tropical regions where agricultural burning is prevalent. The simulations also took into account the solar absorptive properties of black carbon, the first time this effect had been modeled.

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The simulations showed that biomass aerosols suspended in the clear sky cool the climate by between 0.16 and 0.23 watts per square meter, while black carbon from fossil fuels heats the climate by between 0.16 and 0.20 watts per square meter. Also, sulfate aerosols cool the atmosphere by between 0.53 to 0.81 watts per square meter. The sum of the cooling effects ranges between 0.35 and 0.65 watts per square meter. (To place these figures in perspective, about 340 watts per square meter of



solar radiation reaches Earth's atmosphere daily.)

Chuang's research then moved to the vastly more complex task of modeling the indirect effects of anthropogenic sulfate and carbonaceous aerosols through their interaction with clouds. These simulations indicated that the indirect effects of aerosols are greater than the direct effects. The simulations also showed that aerosols can mask the warming effects of greenhouse gases, at least in regions with high pollution levels.

Aerosol-Cloud Interactions

The simulations estimated that aerosols acting as CCN cool Earth by about 1.85 watts per square meter, with

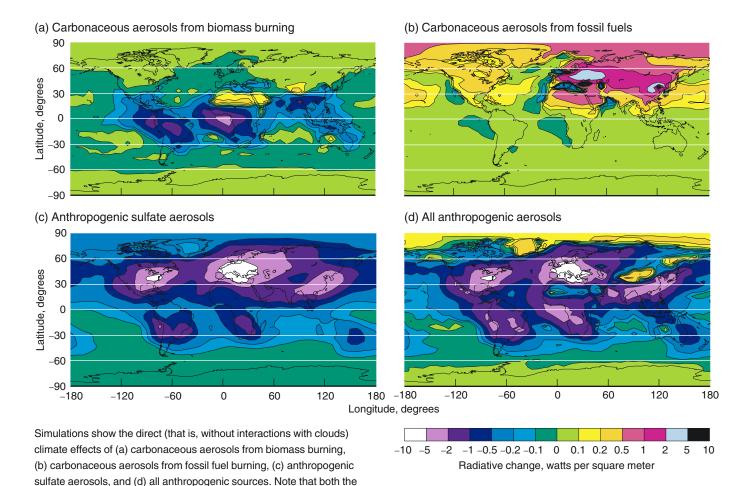
fossil fuel and biomass burning release sulfur dioxide, which later

oxidates to sulfate.

0.30 watts per square meter associated with anthropogenic sulfate, 1.16 watts per square meter associated with carbonaceous aerosols from biomass burning, and 0.52 watts per square meter associated with carbonaceous aerosols from fossil fuel combustion. While concentrations of anthropogenic carbonaceous aerosols are about equal in the Northern and Southern hemispheres, aerosols of anthropogenic sulfates are more prominent in the Northern Hemisphere.

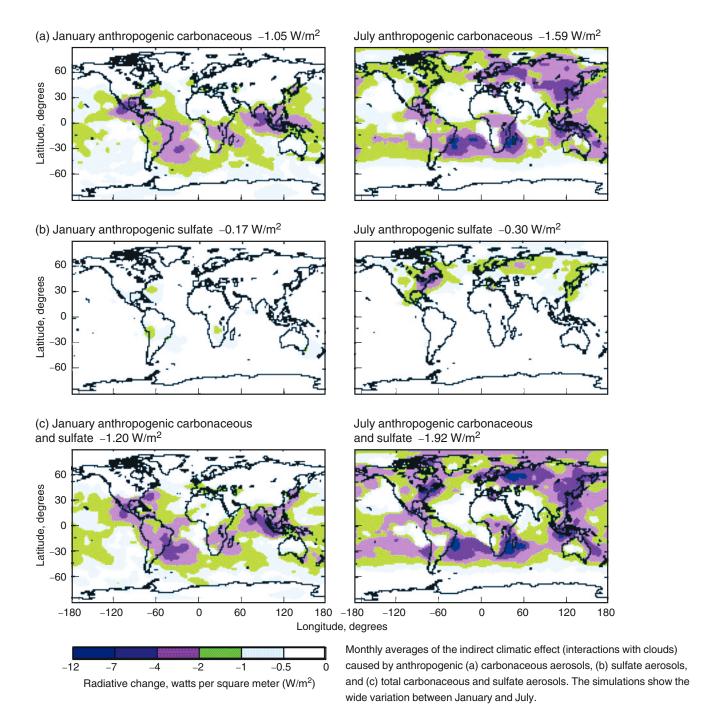
Also, the simulations showed that concentrations of aerosols vary with the seasons. The global average of indirect effects by anthropogenic aerosols is greatest in April through June, a period when biomass of savanna and forested areas is burned in the topics. The indirect cooling effect is highest in May, with 2.4 watts per square meter.

Chuang also addressed how black carbon absorption affects solar energy in clouds. She found that including this absorption does not decrease the overall cooling effect by more than 0.07 watts per square meter on a global scale, but that locally, it can decrease the cooling effect by as much as 0.7 watts per square meter in regions that have significant black carbon emissions. The model shows that if the effect of black carbon absorption in clouds is not included, the indirect cooling effect by carbonaceous aerosols may be overestimated by up to



25 percent in regions where black carbon emissions are significant.

The Livermore assessments were based on a three-dimensional general circulation model called Community Climate Model-1 (CCM-1), which was developed by the National Center for Atmospheric Research in Boulder, Colorado. General circulation models predict global changes that result from changing concentration of gases by dividing the global atmosphere into tens of thousands of boxes and using the equations describing motion, energy, and mass to predict the changes in climate. Chuang linked CCM-1 to GRANTOUR, a three-dimensional global chemistry code for the



troposphere, which was first developed at Livermore in the late 1980s to simulate the concentration and distribution of aerosols and their gaseous precursors.

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"Both GRANTOUR and CCM-1 were developed more than a decade ago," notes Chuang. "They lack advanced physics and modeling techniques that prevent us from exploring in greater detail the interrelationship between aerosols, clouds, and climate variation.

We believe this interrelationship is the leading source of uncertainty in predicting future climate change."

New Era of Modeling

The research team's goal is to move to what Chuang describes as a "new era of modeling" that will link the most advanced atmospheric chemistry and climate codes. To that end, last year Chuang and her colleagues added improvements to Livermore's integrated, massively parallel atmospheric chemical transport (IMPACT) code so that it better represents aerosol chemistry and runs faster on multiparallel supercomputers. IMPACT, which was previously applied by Livermore researchers to global ozone calculations, includes both the stratosphere and troposphere and uses databases of monthly averaged emissions compiled by scientists and government agencies worldwide to treat global chemistry processes.

Aerosols: A Short Primer

Aerosols are concentrations of exceedingly minute particles suspended in the atmosphere. Aerosol particles range in size from 0.01 micrometer (millionth of a meter) to several tens of micrometers in diameter. Particles generated by pollution tend to be less than a millimeter in diameter.

The particles enter the atmosphere from many different natural and anthropogenic (human activity-related) sources. For example, nature generates sulfate aerosols from volcanoes, salt aerosols from sea spray, dust aerosols from desert areas, and carbonaceous aerosols formed from volatile organic compounds emitted by plants.

A growing fraction of aerosols are byproducts of human activities, as seen in the ubiquitous hazes that persist in the industrialized regions of the world. Anthropogenic aerosols include sulfuric acid, soot and smoke from the burning of fossil fuels in factories, vehicles, power plants, cookstoves, and fireplaces. The burning of forests and grasslands to clear them for farming is another source of carbonaceous aerosols. (Although dust is typically considered a natural source of aerosols, human activities such as farming or erosion caused by changing land use also kick large amounts of dust into the atmosphere.)

Aerosols have a significant effect on climate. Whereas greenhouse gases trap the Sun's heat, thereby warming Earth's atmosphere and surface, aerosols mainly reflect solar radiation, a phenomenon called the aerosol direct effect. By reducing the amount of solar energy reaching the Earth's surface, aerosols serve as agents of climate cooling.

Aerosols also cool the climate indirectly, by changing the properties of clouds, which cool Earth by reflecting solar radiation back to space. (Of the daily average of about 340 watts per square meter of solar radiation that reaches the atmosphere, clouds reflect about 45 watts per square meter.) Although commonly thought of as pristine sources of water, clouds could not form without aerosol particles (natural or anthropogenic) acting as cloud condensation nuclei, which are sites on which water droplets can condense.

Reflecting Sunlight, Modifying Rainfall

Higher concentrations of aerosols in the atmosphere lead to the formation of clouds with water content spread over many more particles. Clouds with smaller, more numerous droplets have a larger surface area and therefore reflect up to 30 percent more sunlight, a phenomenon called aerosols' first indirect effect. What's more, the smaller water droplets in the cloud fall more slowly, thereby prolonging the lifespan of the cloud and strengthening its cooling effect. This second indirect effect is believed to be changing rainfall patterns in populated regions worldwide.

Complicating the scientific understanding of aerosols' climatic effects are recent satellite observations revealing that aerosols of black carbon from biomass and fossil fuel burning can absorb sunlight in the atmosphere, thereby increasing the warming effect of greenhouse gases. Satellite observations have also revealed that the absorption of heat by soot can evaporate cloud droplets and thus reduce the presence of clouds. This phenomenon, called the aerosol semidirect effect, is particularly prevalent over heavily polluted areas.

Taken together, all of the direct and indirect effects of aerosols are believed to increase Earth's albedo (percentage of sunlight reflected), thereby cooling the surface and offsetting the warming effects of greenhouse gases by 25 to 50 percent globally (and even much more in some areas).

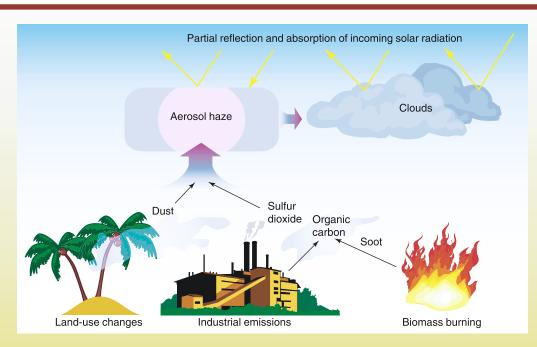
However, aerosols' climatic effects cannot be simply compared to those of greenhouse gases because they are distributed in time and space far differently. For example, greenhouse gases are well mixed in the atmosphere and have a lifetime of up to 100 years. In contrast, aerosols suspended in the troposphere (lower atmosphere) last only about a week before they are removed by winds and rain. (The exception is the injection of sulfates into the stratosphere, or upper atmosphere, where they can remain for a few years. The global cooling observed following large volcanic eruptions, such as

With the recent revisions, IMPACT can simulate the complicated reactions involving sulfate aerosols that are formed from sulfur dioxide generated by power plants and biomass burning. The code can also account for other sources of sulfates, including the production of dimethylsulfide by plankton, sulfur dioxide by volcanoes, and hydrogen sulfide by soils, forests, and crops. The new version of IMPACT also predicts the concentrations of black carbon and

other carbonaceous compounds, dust, and sea salt as well as their seasonal variations.

To better represent the ever-changing size distribution of aerosol particles, Chuang is adapting an aerosol microphysics module developed at Brookhaven National Laboratory. The module simulates aerosol dynamics through complicated nucleation, growth, and transport processes by tracking the lower order moments of an aerosol size distribution in space and time.

Chuang plans to link IMPACT and the new microphysics module with the Community Climate Model-3, or CCM-3, the fourth-generation model developed by the National Center for Atmospheric Research. This climate model allows more realistic and higher resolution simulations of aerosol effects on regional climate. For example, it can show how aerosols are transported to different regions by strong winds and removed by rainfall.



Clouds could not form without aerosol particles (natural or anthropogenic) acting as cloud condensation nuclei or sites on which water droplets can condense. Anthropogenic emissions increase aerosol concentrations and result in clouds with smaller and more numerous droplets. These clouds have a larger albedo (percentage of reflected sunlight) and a longer lifetime, and thus they reflect more sunlight back into space.

that of Mount Pinatubo in the Philippines in 1991, provides dramatic evidence for the climatic influence of aerosols.)

Also, many anthropogenic aerosols are localized and occur near or downwind from their sources, such as power plants, factories, and large urban populations. As a result, most aerosols are found in the Northern Hemisphere, where most industrialized nations are located.

Instrument Data, Models Aid Understanding

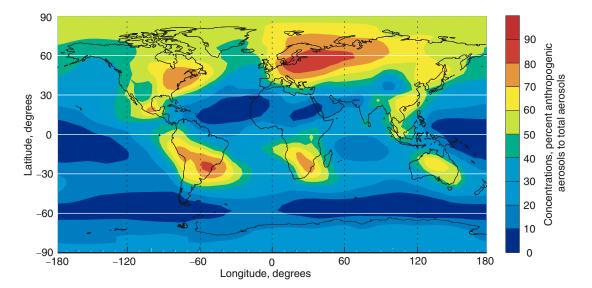
To accurately study aerosol distribution and composition requires continuous observations from instruments located on satellites and aircraft as well as ground-based field stations. Data from these instruments, combined with numerical models that mimic the formation of aerosols and their interactions with clouds, have led to a much greater understanding of how and to what degree aerosols influence

climate. Lawrence Livermore scientists have been among the leaders in developing these models.

Aerosol research marked a turning point in January 2002, when more than 50 leading American atmospheric scientists (including Livermore's Catherine Chuang), together with representatives from federal agencies, met to explore ways to achieve breakthroughs in understanding and modeling aerosols' role in climate change. The meeting led to the formation of a national Aerosol Climate Interactions Program supported by several federal agencies. The program's goals are to more accurately measure the sources, distribution, and properties of aerosols and their influence on climate; to more completely model the processes that govern aerosols' distributions and climatic effects; and to better quantify the relative importance of aerosols and greenhouse gases in global warming, including the effects on regional climates.

This simulation, using IMPACT, shows the percentage of concentrations (averaged on an annual basis) from all anthropogenic sources of aerosols.

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Chuang notes that CCM-3 is typically used by research centers at 300-kilometer resolution. Such coarse resolution limits the code's usefulness because it does not adequately represent topographic features that strongly influence surface temperature and precipitation. Much finer resolutions are required to examine regional climate change and the transport of aerosols through the atmosphere. Chuang notes that a Livermore team headed by Philip Duffy has simulated the effects of increased



greenhouse gases by using CCM-3 at 50-kilometer resolution to obtain the finest resolution of global warming performed to date. (See *S&TR*, July/August 2002, pp. 4–12.)

Putting Everything Together

With all the modeling elements in place, Livermore atmospheric scientists will be able to simulate early next year the global and regional climate changes caused by both aerosols and major greenhouse gases. "With more complete chemistry and physics in our models, we hope to have more accurate answers about how human activities are affecting our climate," Chuang says.

Ultimately, she says, realistic climate models—augmented by other data—provide the only viable approach for determining how aerosols are changing the planet's climate and for assessing the effects of future emissions. "Models are the only tools for making predictions about climate change so that we can help policymakers arrive at the most informed decisions for responding to changes in the environment."

She notes that at first glance, it might seem that aerosols are a positive element because they tend to counter the effects of global warming. However, purposely allowing a greater buildup of aerosols to offset global warming would lead to greater health and ecological damage. Aerosols that affect climate are associated with air pollution and acid rain, lower visibility, and decreased agricultural production.

The results from the advanced Livermore simulations will surely help society as it decides to manage air pollution, global warming, changing rainfall patterns, and the unavoidable effects on human health and society.

-Arnie Heller

Key Words: aerosols, biomass, black carbon, cloud condensation nuclei (CCN), Community Climate Model (CCM), dimethylsulfide, global warming, GRANTOUR, IMPACT, National Center for Atmospheric Research, soot, sulfate.

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